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APPLICATION NUMBER: 60/374,512

FILING DATE: April 23, 2002

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# PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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## INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
LORRIE JIM ROD	HUNT McKINLEY McELROY	Delta, Canada North Vancouver, Canada

☐ Additional inventors are being named on the \_\_\_\_\_ separately numbered sheets attached hereto

## TITLE OF THE INVENTION (500 characters max)

REMEDIATION OF SOILS CONTAMINATED WITH POLYCHLORINATED BIPHENYLS (PCBs) AND OTHER ORGANOHALOGENS BY SIMULTANEOUS PCB EXTRACTION AND DESTRUCTION

## Direct all correspondence to: CORRESPONDENCE ADDRESS

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## ENCLOSED APPLICATION PARTS (check all that apply)

☒ Specification Number of Pages 10

☐ Drawing(s) Number of Sheets

☐ Application Data Sheet. See 37 CFR 1.76

☐ CD(s), Number

☐ Other (specify)

## METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☐ Applicant claims small entity status. See 37 CFR 1.27.

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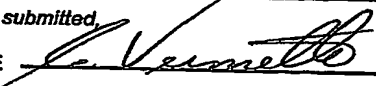
FILING FEE AMOUNT (\$) \$160.00

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

☒ No.

☐ Yes, the name of the U.S. Government agency and the Government contract number are: \_\_\_\_\_

Respectfully submitted,

SIGNATURE 

TYPED or PRINTED NAME Clifford W. Vermette

TELEPHONE (604) 331-0381

Date

REGISTRATION NO. (if appropriate) 30,018

Docket Number: 1635-255

## USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

This collection of information is required by 37 CFR 1.51. The information is used by the public to file (and by the PTO to process) a provisional application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the complete provisional application to the PTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, Washington, D.C. 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Box Provisional Application, Assistant Commissioner for Patents, Washington, D.C. 20231.

# FEE TRANSMITTAL for FY 2002

Patent fees are subject to annual revision.

TOTAL AMOUNT OF PAYMENT (\$) 160.00

## Complete if Known

Application Number  
Filing Date  
First Named Inventor **Lorrie Hunt**  
Examiner Name  
Group Art Unit  
Attorney Docket No. **2313-100**

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## FEE CALCULATION

### 1. BASIC FILING FEE

Large Entity Code (\$)	Small Entity Code (\$)	Fee Description	Fee Paid
101 740	201 370	Utility filing fee	
106 330	206 165	Design filing fee	
107 510	207 255	Plant filing fee	
108 740	208 370	Reissue filing fee	
114 160	214 80	Provisional filing fee	160.00

SUBTOTAL (1) (\$) 160.00

### 2. EXTRA CLAIM FEES

Total Claims	Extra Claims	Fee from below	Fee Paid
3	-20** =	X	
Independent Claims	-3** =	X	
Multiple Dependent			

Large Entity Code (\$)	Small Entity Code (\$)	Fee Description
103 18	203 9	Claims in excess of 20
102 84	202 42	Independent claims in excess of 3
104 280	204 140	Multiple dependent claim, if not paid
109 84	209 42	** Reissue independent claims over original patent
110 18	210 9	** Reissue claims in excess of 20 and over original patent

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## FEE CALCULATION (continued)

### 3. ADDITIONAL FEES

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
105 130	205 65	Surcharge - late filing fee or oath	
127 50	227 25	Surcharge - late provisional filing fee or cover sheet	
139 130	139 130	Non-English specification	
147 2,520	147 2,520	For filing a request for ex parte reexamination	
112 920*	112 920*	Requesting publication of SIR prior to Examiner action	
113 1,840*	113 1,840*	Requesting publication of SIR after Examiner action	
115 110	215 55	Extension for reply within first month	
116 400	216 200	Extension for reply within second month	
117 920	217 460	Extension for reply within third month	
118 1,440	218 720	Extension for reply within fourth month	
128 1,960	228 980	Extension for reply within fifth month	
119 320	219 160	Notice of Appeal	
120 320	220 160	Filing a brief in support of an appeal	
121 280	221 140	Request for oral hearing	
138 1,510	138 1,510	Petition to institute a public use proceeding	
140 110	240 55	Petition to revive - unavoidable	
141 1,280	241 640	Petition to revive - unintentional	
142 1,280	242 640	Utility issue fee (or reissue)	
143 480	243 230	Design issue fee	
144 620	244 310	Plant issue fee	
122 130	122 130	Petitions to the Commissioner	
123 50	123 50	Processing fee under 37 CFR 1.17(q)	
126 180	126 180	Submission of Information Disclosure Stmt	
581 40	581 40	Recording each patent assignment per property (times number of properties)	
146 740	246 370	Filing a submission after final rejection (37 CFR § 1.129(a))	
149 740	249 370	For each additional invention to be examined (37 CFR § 1.129(b))	
178 740	278 370	Request for Continued Examination (RCE)	
169 900	169 900	Request for expedited examination of a design application	
Other fee (specify)			

\*Reduced by Basic Filing Fee Paid

SUBTOTAL (3) (\$)

## SUBMITTED BY

Name (Print/Type) **Clifford W. Vermette** Registration No. **30,018** Telephone **(604) 331-0381**  
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REMEDICATION OF SOILS CONTAMINATED WITH POLYCHLORINATED BIPHENYLS  
(PCBS) AND OTHER ORGANOHALOGENS BY SIMULTANEOUS PCB EXTRACTION  
AND DESTRUCTION

5 FIELD

The present invention relates to a method of simultaneously extracting and destroying polychlorinated biphenyls (PCBs) in contaminated solids, such as soil.

10 BACKGROUND

15 The existence of large numbers of sites with soil extensively contaminated by organohalogens, especially PCBs, requires economical cost effective treatment methods. Although incineration has been shown capable of destroying PCBs this form of PCB treatment has been implicated in dioxin and chlorinated dibenzofuran emissions and has been banned for use in certain countries such as Australia and Japan (References 1,2). Both furan and dioxins are considered to be highly toxic (carcinogenic). As such, there is a need for cost effective PCB decontamination methods, including soil decontamination, which do not involve PCB incineration.

20 Getman et al in U.S. patent 6,049,021 describe remediation of soil contaminated with PCBs. This patent describes the destruction of PCBs in soil using a variety of methods combining the following basic elements:

1. PCB extraction of soil by liquid ammonia.

2. Dissolution of sodium metal into PCB-contaminated liquid ammonia.
3. Destruction of PCB in liquid ammonia by dissolved sodium metal.

Although this technique clearly shows high destruction of PCB in soil it suffers from the following problems:

- Very long reaction times e.g. >30 minutes (see Example 2).
- Need to refrigerate ammonia with soil with stirring before addition of sodium metal (see Example 4).
- Need to operate with hazardous pressurized anhydrous ammonia gas in a stirred vessel (see Example 2).
- Extremely high ammonia dose on soil e.g. 9 litres ammonia per kilogram of soil (see Example 2).
- Generation of ammonia containing residual wastes "filtrates" (see Example 2).
- Awkward temperature cycling between 0 °C and 20 to 40 °C (see Example 4) or -78°C (see Example 3).
- Awkward, time consuming, multiple soil extractions with ammonia before addition of sodium metal (see Example 4).

United States patent 5,228,921 to Peterson describes a method for extracting organohalogens from organohalogen contaminated solids such as PCBs from PCB contaminated soils. United States patent 5,376,182 to Everett et al describe PCB extraction from PCB contaminated soil with ultrasound at 10 to 60 kilohertz frequency. Although these extraction methods successfully remove PCBs from soil, they do not destroy the PCBs.

PCT application WO 02/22252 A1 to Collings describes ultrasonic destruction of PCBs in a one-step process. However processing times are excessive (e.g. 60 minutes) and PCB destruction efficiency is low (e.g. 75%) (page 10, lines 20-25).

Eco Logic at web page [www.eco-logic-intl.com](http://www.eco-logic-intl.com) in a brochure dated April 2001 and entitled "The TORBED/GPCR combination for Soil, Sediment and Sludge Treatment" describe a multi-step process for removal and destruction of PCBs in solids such as soils as follows:

1. High temperature (e.g. 600°C) thermal desorption of PCBs from soils by volatilization.
2. High temperature (e.g. 875°C) gas phase reduction of volatilized PCB exhaust gas from step 1 with a reducing gas such as hydrogen.
3. Scrubbing of exhaust gas from step 2 to recover toxic and/or corrosive gases such as hydrogen chloride produced from reduction of PCBs.

4. Compression and/or storage of scrubbed exhaust gas from step 3.

5 5. Incineration and/or recycling of scrubbed exhaust gas from step 4 to steps 1 and/or 2 respectively.

Although the Eco Logic method clearly destroys PCBs in soils it suffers from the following problems:

- Generation of toxic and/or corrosive exhaust gas (e.g. hydrogen chloride) and spent scrubber solutions.
- Use of potentially explosive hydrogen gas at high temperature.
- Five or more processing steps.
- Two energy intensive, high temperature processing steps.

There is a need for a low temperature process which can quickly extract and destroy PCBs in one step thereby reducing the number of processing steps, the size of equipment and their cost.

## 25 SUMMARY OF THE INVENTION

The current invention relates to the simultaneous extraction and destruction of PCBs in contaminated solids, such as soil. The current invention quickly extracts and destroys PCBs by combining the contaminated soil with an alkali metal,

such as sodium, as well as an inert alkali-metal liquid carrier, such as kerosene. The combination is then placed in a sealed sonication chamber to effect melting of the alkali metal, which causes simultaneous extraction of PCB from the contaminated soil into the liquid carrier and destruction of PCBs by the molten alkali metal. A method of stripping the inert alkali-metal liquid carrier is then employed, resulting in remediation of the contaminated soil.

In contrast to the prior art, the use of this method allows remediation of solids, such as soil, in one reaction chamber using sonication, and at relatively low temperatures, which avoids the highly toxic emissions caused by high temperature incineration.

One advantage of the current invention is the use of kerosene as an extractant resulting in a significant reduction in the amount of extractant required.

#### DETAILED DESCRIPTION

The following remediation method can be applied to solids, such as soil, contaminated by organohalogens, especially PCBs:

1. Air-dry and sieve PCB contaminated soil.
2. Combine sieved, air-dried PCB contaminated soil from step 1 with an alkali metal (e.g. sodium) in an essentially inert alkali-metal liquid carrier (e.g. kerosene or other hydrocarbon) and place inside a sealed sonication chamber.



3. Heat the sonication chamber from step 2 to effect melting of the alkali-metal (e.g. sodium) and apply sonication to effect simultaneous extraction of PCB from the contaminated solids into the liquid carrier and destruction of extracted PCBs by molten alkali metal (e.g. sodium).

4. Discharge the sonication chamber reaction product from step 3 and use any one of the non-limiting soil/liquid carrier separation, alkali hydroxide (e.g. sodium hydroxide) by-product neutralization methods described below.

5. Recycle the treated soil.

Referring to step 4, a variety of non limiting-techniques for stripping the inert alkali-metal carrier (e.g. hydrocarbon such as kerosene) from the soil together with soil recycling include the following:

- For coarse-grained soils, addition of water (the same or higher volume than the hydrocarbon oil) followed by vigorous mixing in the sonic generator chamber (or in a separate vessel) results in water wetting of the solid phase. When the agitation is stopped, the oil floats on the water phase and can be recovered by decantation. Water addition/mixing followed by settling and oil decantation will be the primary stage of oil stripping in most cases.
- More commonly the soil in the water phase after primary oil decantation will retain a significant amount of oil and

require treatment by froth flotation to achieve the required oil content for recycling.

- When stringent criteria ( $<20$  ppm) apply to the oil content of recycle soil, oil sorption by coarse granular activated carbon can be used to achieve very low residual oil levels.

- The oil recovered by decantation will normally contain a portion of the fine-grained solids from the treated soil. Depending on the nature of the soil being treated, methods to recover solids from the recycle oil phase include secondary water scrubbing with a surfactant, centrifugation or filtration.

- Removal of water from treated soil can be achieved by simple drainage for coarse-grained soil. For finer material vacuum or pressure filtration will provide a handleable material for recycle.

The methods described above are either standard practice or slight variants of standard mineral processing operations (references 3,4).

Modern practice in solvent extraction also provides examples of technology for removal of fine solids ("crud" in industry jargon) from the organic phase which typically contains  $>50\%$  hydrocarbon oil as diluent as well as the oil soluble extraction reagent.

During the water contact /oil separation operations, sodium hydroxide (NaOH) is formed from any residual sodium metal and by hydrolysis of the sodium-aromatic hydrocarbon bond formed during the dechlorination reaction. Sodium hydroxide is known in the mineral processing industry as a (water) wetting agent for many minerals, and may assist in the oil removal operation. However, neutralization of sodium hydroxide will be required prior to discharge of process water. This can be achieved by a variety of non-limiting methods including:

- Injection of carbon dioxide gas into the aqueous phase to convert sodium hydroxide into sodium carbonate or bicarbonate
- Addition, under pH control, of sulfuric acid to form neutral sodium sulphate

The following example illustrates the effectiveness of the invention:

#### Example 1

1. PCB-contaminated soil was air dried and sieved to -6 mesh.
2. Two kilograms of soil from step 1 was combined with 0.6 litres of commercially available kerosene (i.e. a 0.3 litre kerosene to 1 kg soil weight ratio) and 45 grams of solid sodium metal in a 3.2 litre grinding media-free sonication chamber attached to a 20 kilowatt sonic driver.
3. The sealed sonication chamber was mounted on the 20 kilowatt sonic driver and was heated to 115°C using heat from a propane torch. The sonic chamber heating jacket was filled half-

way with ethylene glycol antifreeze to aid in heat transfer to the sonic chamber ingredients. Once the desired temperature was reached (e.g. alkali metal melting point or higher), sonication was commenced.

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As an example, the sonication temperature was 114°C at the end of a 6 minute run. The following table illustrates PCB destruction as a function of time using the above approach on a soil with an initial PCB content of 424 ppm (micrograms/gram):

10

Sonication Time in minutes	PCB ppm in soil	% PCB destruction
0	424	0
1	12.7	97.0
2	8.4	98.0
5	2.2	99.5
6	2.5	99.4

A 99.5% destruction of 424 ppm PCB contaminated solid with a 45 g. sodium to 2000 g soil dose ratio was achieved in 5 minutes.

15 Using kerosene as an extractant required approximately 0.3 liters of kerosene to one kilogram of soil. This ratio represents a 30 fold reduction in the volume of extractant required as compared to the ammonia extractant of the Getman patent.

20

## References

1. Costner, Pat et al. 1998. "Technical Criteria for the Destruction of Stockpiled Persistent Organic Pollutants". Third Meeting of the Intersessional Group Intergovernmental Forum on Chemical Safety. Yokohama, Japan. December 1-4, 1998. Available at [www.eco-logic-intl.com](http://www.eco-logic-intl.com)
2. "Survey of Currently Available Non-Incineration PCB Destruction Technologies". 2000. United Nations Environment Programme. August. Available at [www.eco-logic-intl.com](http://www.eco-logic-intl.com)
3. Taggart, Arthur F. 1945. Handbook of Mineral Dressing. John Wiley and Sons Inc. (New York)
4. Gaudin, A.M. 1957. Flotation. McGraw-Hill Book Co. Inc. (New York)